PROCEEDINGS
OF THE
NATIONAL ACADEMY OF SCIENCES
Volume 10 NOVEMBER 15, 1924 Number 11

THE SECONDARY ELECTRONS PRODUCED BY HARD X-RAYS IN LIGHT ELEMENTS

By Hugo Fricke and Otto Glasser

Department of Bio-Physics, Cleveland Clinic Foundation, Cleveland, Ohio

Communicated, October 10, 1924

A. H. Compton[1,2] has recently proposed a theory for the scattering of X-rays which very successfully explains many of the phenomena which depend on the interaction between light atoms and hard X-rays. A corollary of this theory which we shall test here consists in the prediction and quantitative description of a special type of secondary electrons which result from the passage of X-rays through matter. The actual existence of these electrons which Compton terms *recoil-electrons* has been demonstrated by C. T. R. Wilson[3] and W. Bothe,[4,5] according to whom the range and number of the recoil-electrons produced by X-rays of varying hardness is in fair agreement with Compton's theory.

In the following investigation we shall present an experimental determination of that fraction of the ionization of X-rays in air for which the recoil-electron emission is responsible, offering thereby an indirect determination of the absorption due to the recoil-electron emission, the value for which is given by Compton's theory.

Our experimental method consists in a comparison of the ionization currents produced by X-rays in very small ionization chambers made of different light elements and their salts. Our calculation of the experimental results is based on the following process of reasoning: X-rays produce in matter two types of secondary electrons which are entirely responsible for the ionization produced by the rays. For the case of hard X-rays and light elements, in accordance with Einstein's equation the electrons of one of these types which are due to the photo-electric effect of the rays have the energy $h\nu$; and per gram electron and per unit energy radiation their number is proportional to the third power of the atomic number (or in the case of a chemical compound $a_1N_1 + a_2N_2 - - -$ the effective atomic number

$$N_{\text{eff}} = \sqrt{\frac{a_1N_1^4 + a_2N_2^4 + - - -}{a_1N_1 + a_2N_2 + - - -}}$$

The electrons of the second type are due to the scattering process and according to Compton their velocity and number per gram electron are independent of the atomic number. In passing through matter, these electrons suffer absorption, stopping and scattering; and in the case of light elements all these effects per gram electron are independent of the atomic number. (Compare a discussion by Lenard.[6])

From the above it is easily derived by a geometrical method that the ionization current in an ionization chamber which is small as compared to the path of the photo-electrons in the air of the chamber can be expressed in arbitrary units by

$$I = \frac{\mu_1 N_{\text{eff}}^3}{V_1} + \frac{\mu_2}{V_2} \qquad (1)$$

in which $\mu_1 N_{\text{eff}}^3$ is the coefficient of photo-electric absorption (per gram-electron) of the rays in the material of the chamber and μ_2 is the coefficient of recoil-absorption (per gram-electron). V_1 and V_2 express respectively the number of volts which are used in producing one ion for the case of a photo-electron and for that of a recoil-electron. $\mu_1 N_{\text{eff}}^3/V_1$ represents the ionization due to the photo-electrons from the wall of the chamber; μ_2/V_2 represents the ionization due to the recoil-electrons.

Our experiments will furnish a direct determination of the ratio

$$\frac{\mu_1 N_{\text{eff}}^3/V_1}{\mu_2/V_2} \qquad (2)$$

the value of which can also be calculated from Compton's theoretical value for the coefficient of recoil-absorption and from known experimental data.

In our experiments we have used two types of radiation. Although as will be shown, these are very nearly homogeneous, they are not strictly so and we must, therefore, consider how to determine their effective wave-length. μ_1 in (2) is proportional to the third power of the wave-length. V_1/V_2 and according to Compton's[1] theory also μ_2 are approximately independent of the wave-length. The ratio (2), therefore, varies nearly as the third power of the wave-length; consequently the correct effective wave-length can be obtained by an absorption experiment if the absorber is so heavy that its absorption is solely of the photo-electric type and the absorption of the intensity indicator is independent of the wave-length. We have used silver as an absorber and a small graphite ionization chamber as an indicator. For the range of wave-lengths with which we shall deal (.20 $\AA U$ to .10 $\AA U$) the absorption of graphite is very nearly independent of the wave-length; at .10 $\AA U$ it is solely due to the recoil-electron emission while at .20 $\AA U$ about half is due to the photo-electric effect.

The softer radiation which we have employed is defined by the following characteristics: Potential 100 KV (as determined by spark gap.) Tungsten target, Filter: .43 gr. tungstic acid + .133 gr. Copper + .27 gr. Aluminum per sq. cm. Coefficient of mass absorption in Silver 4.10, in Copper 1.25. Effective wave-length (according to Allen[7] and Richtmyer[8]) $\lambda = .180\ \AA U$ by Silver, $\lambda = .183\ \AA U$ by Copper.

The homogeneity of the radiation is established by the linearity of an absorption curve (Copper) together with the fact that the effective wave-length of the radiation is (within one per cent) equal to the critical K-absorption wave-length of tungsten ($\lambda = .1783\ \AA U$).

The harder radiation employed was defined by the following characteristics: Potential 200 KV, Tungsten target, Filter: .547 gr. Silver + .72 gr. Copper + .27 gr. Aluminum per sq. cm. Coefficient of mass absorption in Silver 1.39, in Copper .354. Effective wave-length by Silver $\lambda = .115\ \AA U$[7], by Copper $\lambda = .106\ \AA U$.[7,8]

This radiation is not as homogeneous as the first type; however as a rigorous calculation shows to a good approximation it is equivalent to a homogeneous radiation of the wave-length .115 $\AA U$.

A description of the details of our experimental technic has been given in other papers[9,10]. A series of different sizes of ionization chambers—cylindrical and rectangular—have been used, the volumes ranging from $^1/_2$ cc. to 9 cc. The important feature of the construction of these chambers is that the ionized air volume is enclosed only by the material under investigation.

TABLE I

Conditions: 100 KV, .43 gr. tungstic acid + .133 gr. Cu + .27 gr. Al per sq. cm.
$\mu/\rho_{Ag} = 4.10$; $\mu/\rho_{Cu} = 1.25$; $\lambda = .180\ \AA U$
Volume of ionization chamber: 1.039 cc.

MATERIAL	COMPOSITION	N	$I_{obs.}$	$I_{calc.}\left(.78 + .22\frac{N^3}{6^3}\right)$	$\Delta\%$
Graphite	C	6	1.00	1.00	0
Urea	NH_4CO	6.25	1.06	1.03	+3
Maleic Acid	$C_2H_2O_2$	7.11			
Ammonium Nitrate	NH_4NO_3	7.47			
Sodium Nitrate	$NaNO_3$	8.85	1.44	1.49	−3
Magnesium*	Mg	12	2.50	2.54	−1
Aluminum	Al	13	3.1	3.0	+3
Potassium Nitrate	KNO_3	14.3	3.8	3.8	0
Sulphur**	S	16	4.8	4.9	−2
Copper	Cu	29	26.0	25.6	+2

* The Magnesium was obtained from the American Magnesium Corporation, Niagara Falls, N. Y. The purity was given as 99.955%. The Aluminum (99.5% pure) was obtained from the Aluminum Company of America, Pittsburgh, Pa. We wish to thank these corporations for their kind help in furnishing these materials.

** The inner surface of the sulphur chamber was covered by a thin layer of graphite.

Among the many materials which have been tested, only those given in table I could be utilized. They must be stable, non-hygroscopic and in the case of the salts, on account of the method by which the chamber is constructed, the melting point must be fairly low. The highest possible purity is essential.

Table I gives the results of a series of actual observations for the softer radiation. The ionization chamber used in this series was of a cylindrical form 1.11 cm. in length, 1.09 cm. in diameter, 1.05 cc. in volume; the electrode was a rod 1.00 cm. in length, .12 cm. in diameter. Under $I_{obs.}$ are given the observed intensities in arbitrary units; under $I_{calc.}$ the intensities calculated by means of equation (1) using

$$\mu_1 N_{\text{eff}}^3/V_1 = .22 N_{\text{eff}}^3/6^3 \text{ and } \mu_2/V_2 = .78.$$

The agreement between the observed and calculated values of I is satisfactory. We may note that some of the materials listed in table I have not been investigated for this particular radiation. However, all materials have been tested under other conditions of radiation and have been found to give readings in agreement with equation (1).

As stated above, a condition for the application of equation (1) is that the free path of a photo-electron in the air of the chamber should be long as compared with the dimension of the ionization chamber. In order to obtain an experimental confirmation of this condition we must show that the relative values of the observed ionization currents of table I are independent of the size of the chamber. For the radiation $\lambda = .180$ $\AA U$, this statement is verified by the observations in table II, which gives relative readings for chambers of different sizes of graphite and aluminum and the intensity per unit volume of chamber, which is seen to be independent of the shape and size of the chamber.* A similar result was obtained for the harder radiation.

In the case of carbon ($N = 6$) the application of equation (1) to our measurements gives

$$\lambda = .180\ \AA U: \qquad \frac{\mu_1 N^3/V_1}{\mu_2/V_2} = \frac{.22}{.78} = .28 \qquad (3)$$

$$\lambda = .115\ \AA U: \qquad \frac{\mu_1 N^3/V_1}{\mu_2/V_2} = \frac{.086}{.914} = .094 \qquad (3)$$

We shall now compare these experimental values with the theoretical values which can be derived from Compton's theoretical value of μ_2 and from other existing data. The coefficient of photo-electric absorption $\mu_1 N^3$ is obtained from existing measurements of the total absorption by subtracting the scattering as given by Compton.[1] Thus, Hewlett's[11] measurements for Oxygen and Allen's[7] measurements for Magnesium give consistent values of μ_1:

$$\mu_1 = .0065 \text{ for } \lambda = .180\ \AA U, \qquad \mu_1 = .00172 \text{ for } \lambda = .115\ \AA U$$

However, it must be noted that by using Allen's[7] measurements for the heavier elements (Aluminum, Sulphur, Iron, Copper) we obtain continuously increasing values of μ_1, this effect probably being due to the nonvalidity of Compton's expression for scattering in the case of the heavier elements.

According to Compton's theory, the value of μ_2 is as follows:

$$\lambda = .180\ \text{Å}U: \quad \mu_2 = .0170, \qquad \lambda = .115\ \text{Å}U: \quad \mu_2 = .0202$$

Using experimental data for the absorption, stopping and ionization (per unit path) of the cathode and β-rays, Lenard[6] has calculated the total number (S) of ions produced by fast electrons from which V can be obtained directly. According to these data, V decreases slowly as the velocity increases. V_1/V_2 would be about $^1/_2$ if the velocity of a recoil-electron is determined by Compton's theory. Lenard, however, has made a serious mistake in his calculation. Since the ionization chambers which have been used in the experimental determination of the ionization of cathode rays have always been narrow in comparison with the path of the rays, the calculation should have employed the true absorption coefficient (α_0) which would be obtained if the rays were not scattered; instead, Lenard has used the absorption coefficient (α), which is obtained by the direct observation of broad bundles of completely scattered rays. An indication of the order of magnitude of α/α_0 may be obtained by Schmidt's[13] theory, according to which α/α_0 is equal to 1.6 for β-rays, its value increasing slightly with decreasing velocity. On account of the incompleteness of the theory by which it is derived this value however is much too uncertain for a calculation to be based on it. As a matter of fact, the recent cloud experiments of C. T. R. Wilson[3] seem to indicate that α_0 is zero; that is that true absorption as defined by Lenard is of negligible importance.

Let us first examine the value of V_1/V_2 if this type of absorption is left out of consideration. In that case V would be calculated from the ratio of the decrease of volt-velocity to the ionization per unit path. It seems hardly possible to obtain the decrease of volt-velocity of fast electrons from the present experimental data, on account of the inconsistencies in the results secured by different observers and especially on account of the uncertainty in their interpretation. We think, however, that there are good reasons for believing that both the decrease of volt-velocity and ionization are exactly stated within the range of velocities with which we are concerned by the Thomson[14]-Bohr[15] theory. According to this theory both the decrease in volt-velocity and the ionization for high velocities vary inversely as the square of the velocity. For smaller velocities the variation is less marked; in these cases, also, their ratio which is equal to V remains constant at first. These laws are in agreement with the exist-

ing experimental data for cathode and β-rays, especially those obtained by W. Wilson,[16] J. L. Glasson,[17] O. V. Baeyer,[18] Widdington,[19] and J. Danysz.[20] The theory has also been verified by the direct observations of the tracks of fast electrons by C. T. R. Wilson.[3]

We therefore conclude that $V_1/V_2 = 1$ if pure absorption as defined by Lenard, can be left out of consideration. Although, as stated before, Wilson's experiments appear to give very definite evidence to this effect, it must be noted that as is well known, it is difficult to account for many other experimental facts without introducing this conception. In any case, however, it seems that the value of V_1/V_2 is approximately 1 and therefore for the present we shall accept this value.

Thus, we obtain as theoretical values:

$$\text{For} \quad \lambda = .180\ \text{Å}U: \quad \frac{\mu_1 N^3/V_1}{\mu_2/V_2} = .38 \qquad \lambda = .115\ \text{Å}U: \quad \frac{\mu_1 N^3/V_1}{\mu_2/V_2} = .085 \tag{4}$$

The agreement between the values given in (3) and (4) is fair although there is a certain discrepancy. Further experimental material is needed however, before an analysis of the reasons for this can be attempted.

TABLE II

Conditions: 100 KV, .43 gr. tungstic acid + .133 gr. Cu + .27 gr. Al per sq. cm. $\mu/\rho_{Ag} = 4.10$ $\lambda = .180\ \text{Å}U$

	IONIZATION CHAMBER DIMENSIONS			ELECTRODE DIMENSIONS			EFF. VOL. OF CHAMBER	CARBON		ALUMINUM	
FORM	LENGTH (CM.)	DIAM. (CM.)	VOL. (CCM.)	LENGTH (CM.)	DIAM. (CM.)	VOL. (CCM.)	(CCM.)	INT.	$\frac{\text{Int}}{\text{VOL.}}$	INT.	$\frac{\text{Int}}{\text{VOL.}}$
Cylinder	2.060	0.558	0.503	2.000	0.119	0.022	0.481	0.92	1.91	2.86	5.95
Cylinder	1.112	1.095	1.050	1.000	0.119	0.011	1.039	1.96	1.88	6.28	6.03
Cylinder	1.540	2.020	4.928	1.400	0.119	0.015	4.913	9.44	1.92	29.40	5.98
Rectang.	3.0;	3.0x.50	4.500	2.4;	2.4x.023	0.133	4.467	8.55	1.91	27.00	6.04

* It is of interest to note that a chamber which is made of a material which has the same effective number as atmospheric air ($N_{eff} = 7.69$) will give the same readings as would be obtained in the same volume of an unlimited airspace. This fact may serve as the foundation for the experimental definition of the unit for the intensity of X-rays, which recently has been introduced in X-ray therapy.[10]

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*THE DIFFUSION OF HYDROGEN INTO AIR, MEASURED BY THE INTERFEROMETER U-GAUGE**

BY CARL BARUS

DEPARTMENT OF PHYSICS, BROWN UNIVERSITY

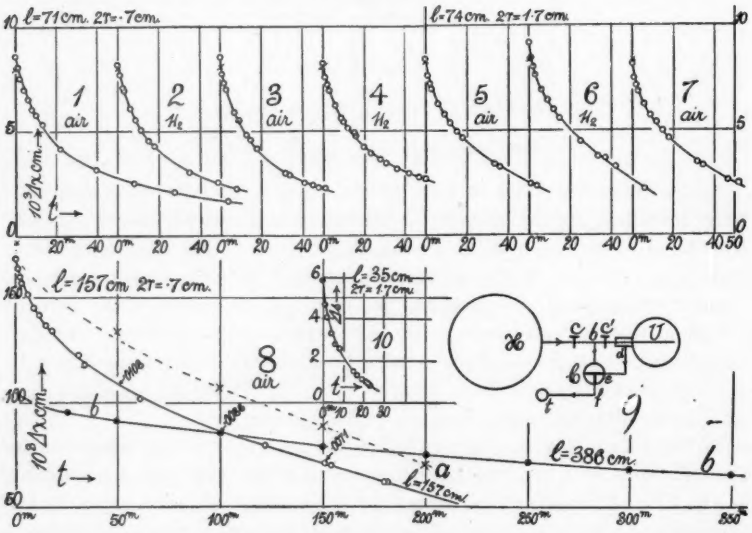
Communicated, September 26, 1924

1. *Introductory.*—The method described heretofore** consists essentially in measuring the pressure at the closed top of a vertical tube, while the charge of hydrogen diffuses out of the bottom. The modified form of apparatus is shown, in plan, in figure 9, where H is the hydrogen reservoir, c and c' stopcocks, U one shank of the U-gauge, t a section of the vertical diffusion tube. C is a threeway cock, by which t may be put in communication with H alone, via *Hbft*, or with U alone, via *Udeft*, as in figure 9, (c' being closed in both cases), alternately. The cock c' is opened incidentally for flooding the closed shank U with hydrogen.

In the diffusion curves, figures 1 to 8, Δx denotes the displacement of the slide micrometer in centimeters, necessary to bring the fringes back to zero at the times t (minutes after filling the tube with hydrogen) given by the abscissas. Δx is proportional to the pressure (p cm. of mercury) at the head of the tube, or $p = g\,\rho_m \cos\Theta.\,\Delta x$, Θ being the angle of incidence of the rays of the interferometer. The length l and diameters $2r$ of tubes are given in each case. Crosses show the initial reading computed from the standard density of hydrogen. In series 1, 3, 5, 7 and 8 the gas at U was air; in series 2, 4, 6, hydrogen. As no advantage was detected for the latter case, it was abandoned.

2. *Observations.*—In the first and second series, the cocks were of about 1 millimeter bore as compared with the tube diameter of 7 mm. In series III and IV the bore of the cocks was reduced to mere crevices, the same diffusion pipe being retained. In series III incidental dislocation of fringes resulting from the H_2 charge at U as it gradually vanishes, increased the diffusion rates spuriously. In series V and VI, retaining the same fine

bore cocks, a wide diffusion tube was substituted. Series V is among the best of the lot. In series VI the commotion due to influx of H_2 produces a condition similar to that in series III. If we omit case III, the ratio of H-pressure at the beginning and end of 50 minutes of diffusion has the mean value 3.19 for the thin (diam. 7 mm.) pipe. Inasmuch as the series VI is not trustworthy, a special series VII was investigated. The attempt was also made to connect C and U by metallic and by glass capillary tubes; but these failed because of the viscosity of the gas. In fact, the U-tube behaved like a closed region with a wandering zero. Omitting case VI, the pressure ratio 3.30, the mean of series V and VII, may be taken as holding the wide diffusion tube (bore 1.7 cm.).



To compute the coefficient of diffusion, a^2, when the escape of hydrogen takes place at the lower mouth only, the equation

$$a = \frac{l}{2\sqrt{\pi t}} \frac{\Delta x_0}{\Delta x} (1 - l^2/24a^2t + l^4/480a^4t^2 - ..) \qquad (1)$$

is available. The series converges slowly, unless t is large and for this reason the time interval $t = 50$ min. was here taken throughout. The pressure ratio at the top is equivalent to $\Delta x_0/\Delta x$, where Δx_0 and Δx are the slide micrometer displacements at the beginning and end of $t = 3000$ sec. For series I, II, IV, we therefore have $l = 71$ cm., $t = 3000$ sec., $\Delta x_0/\Delta x = 3.19$ at 23°, when uncorrected $a' = 1.167$. The terms in

parenthesis are $-.0515$ and $.0032$; whence $a = 1.167 \times .9516 = 1.111$ and the diffusion coefficient $a^2 = 1.234$. For the case of series V, VII obtained with the wide tube, $l = 74$ cm., $t = 3000$ sec., $\Delta x_0/\Delta x = 3.30$ at 22°; whence $a' = 1.258$ and the correction terms $-.0481$ and $+.0028$, respectively. This makes $a = 1.258 \times .955 = 1.201$ and $a^2 = 1.440$. The mean of these values, $a^2 = 1.337$, is about twice as large as the vlaue for air-hydrogen (.66 at 17° C.) usually given. It thus becomes a question to ascertain reasons for this large divergence. It cannot be explained in terms of the capillary discrepancy at U, nor as the result of leakage at the top of the tube for wide variations of diameter give the same result. The effect of rise of the U-surface is slight. Temperature helps but little.

Another feature in the application of equation (1) is the change of a with the lapse of time t, very slowly at first, but finally quite rapidly. Thus if the computation is made for $t = 15$ minutes

Ser. I, II, IV; $2r = .7$ cm., $a = 1.168\ (1 - .1709 + .0350) = 1.009$

Ser. V, VII; $2r = 1.7$ cm., $a = 1.176\ (1 - .1832 + .0703) = 1.008$

so that the data now coincide and also agree with the long tube data below. Convection may have something to do with this as this drop for wide tubes is larger.

3. *Inversion.*—To consider the concomitant diffusion of air into hydrogen let ρ'_0 be the atmospheric density of air and a' the corresponding coefficient, ρ' the density of air at t sec., at x cm. from the end of the tube. Expanding the integral as heretofore, and integrating for the length l of tube

$$\bar{\rho}' = \rho'_0 \left(1 - \frac{l}{2a'\sqrt{\pi t}}(1 - \text{etc.})\right)$$

where $\bar{\rho}'$ is the mean density at t seconds of the air left in the tube.

If we add the corresponding mean density of hydrogen at the same time, the total mean density, $\bar{\rho}$, of the mixed gas in the tube is

$$\bar{\rho} = \rho'_0 \left(1 - \frac{l}{2a'\sqrt{\pi t}}(1 - \text{etc.})\right) + \frac{\rho_0 l}{2a\sqrt{\pi t}}(1 - \text{etc.})$$

Ignoring the correctives temporarily for brevity, the densities may be converted into hydrostatic pressures, $p = gl\rho$, etc.; whence,

$$\bar{p} = p'_0 - \frac{l}{2\sqrt{\pi t}}(p'_0/a' - p_0/a)$$

If we solve this for a, the reduced result is

$$a = \frac{l}{2\sqrt{\pi t}} \frac{(a/a')(p'_0 - p_0) - p_0(1 - a/a')}{p'_0 - \bar{p}} \qquad (2)$$

where $(p'_0 - p_0)/(p'_0 - \bar{p}) = \Delta x_0/\Delta x$

If now $a = a'$, (diffusion of air into H_2 and H_2 into air, identical) the result is equation (1) as used in the above computations with the inclusion of the correctives. If a differs from a' a single equation with two variables $1/a'$ and $1/a$, is implied.

Thus if we accept $a^2 = .66$ or $a = .81$ for example the value of a' computed from series I, II, IV above, would be $a' = 1.126$.

4. *Long Tube.*—Whatever the cause of outstanding differences may be, it will be modified by using the longest tube vertically available in the present installment. This was of length $l = 157$ cm. and diameter $2r = .7$. The fringe displacement is thus about twice as large as the preceding. The data for diffusion are summarized in figure 8, in which the observation was carried on for three hours. After 2 hours the diffusion curve is practically linear at a rate of $\Delta x/\Delta t = 30 \times 10^{-6}$ cm./min. The error of Δx_0 is now again negative as originally, or the correction $\delta x = +.0008$ cm.

Since the diffusion rates are necessarily slower for long tubes larger time intervals t will be desirable to avoid large correctives. The following data taken at 22.8° C. are relevant, a' being uncorrected.

t SEC	a'	CORRECTING FACTOR	a	a^2	$\Delta X_0/\Delta X$ OBSERVED	$\Delta X_0/\Delta X$ COMPUTED
3000	1.270	.799	1.015	1.030	1.57	1.64
6000	1.124	.887	.996	.992	1.96	2.06
9000	1.111	.918	1.020	1.040	2.38	2.50

Thus the mean values $a = 1.010$ and $a^2 = 1.021$ have apparently been brought somewhat nearer the usual tabulated values (.81 and .66); but they are still in marked degree above them. In figure 8*a*, I have drawn the curve which should be observed if $a^2 = .66$, since a varies as $\Delta x_0/\Delta x$, or as $1/\Delta x$, if the correctives are small. This may be assumed to occur for values of t greater than 100 minutes. The mean difference is about .0020 cm. in Δx. The occurrence of a discrepancy so large and resulting from capillary forces is out of the question, as it must have been detected in the large number of density measurements made. One would have to assume that the mercury surface of the gauge in contact with the atmosphere is supported relatively to the closed shank in contact with hydrogen.

5. *Very Long Tube.*—As the mean values of the coefficient a had fallen when the tube length was doubled, the question arises whether a real limit has been reached. Accordingly the tube length was again increased, this time to over five times the original length. To accommodate the new tube, $l = 386$ cm. and $2r = .7$ cm., it had to be placed at a slant, the top being at the influx cock C, figure 9, 94.5 cm. above the floor, while the far end rested in the distance on the floor. Hence for the density measurements, $l = 94.5$ cm., whereas for diffusion measurements $l = 386$ cm. Far from being disadvantageous, this disposition has much to recommend it. In fact the density measurements came out flawlessly at once.

The diffusions are given in part in the graph b, figure 8, and as a whole are also remarkably smooth. At the beginning, however, there is always some irregularity amounting to 3/105 of the initial displacement Δx_0.

From the unusual length of tube, the diffusion proceeds very slowly, so that a long period t is necessary, if the diffusion equation is to converge rapidly. Observations were made throughout about 700 minutes on the same day and were continued on the next day. After 26 hours about 17% of the original hydrogen pressure is still left in the tube.

The constants are as follows, and one may note that there is here no increase of a with t.

l	t	a'	CORRECTION FACTOR	a	
386 cm.	200 Min.	1.413	.821	1.160	Temperature 23.5°
	400 Min.	1.192	.858	1.018	
	600 Min.	1.118	.884	.989	Mean a = 1.002
	700 Min.	1.118	.898	1.004	

The table shows that 200 min. is not sufficient for the practical convergence of the diffusion equation, but that after 400 minutes this may be assumed.

6. *Short Tube.*—By way of contrast diffusion experiments with a short tube were finally made. The dimensions were l = 35 cm. and $2r$ = 1.7 cm. As the fringe displacements are small they were read off directly (s) and then reduced to Δx and the observed data are reproduced in figure 10. In view of the shortness of the tube, the diffusion is rapid and relatively small time intervals suffice to converge the equation, 10–20 minutes being deemed sufficient. The normal density of hydrogen appeared here at once and there is no appreciable capillary correction.

For the diffusion coefficient it is convenient to take the s values directly, and at t = 0 sec., Δs_0 = 5.8, temperature 23.5°

l CM.	t MIN.	Δs	$\Delta s_0/\Delta s$	a'	CORRECTING FACTOR	a
35	10	2.10	2.76	1.120	.933	1.045
	20	1.05	5.50	1.568	.967	1.516

The diffusion thus increases for some reason, probably thermal convection, in the lapse of time.

7. *Conclusion.*—If finally we compare the values of a (diffusion coefficient a^2) obtained in the different tube lengths and diameters (including the quill tube graph in the preceding paper), and use the smallest time interval t compatible with a convergent equation, the results are (22°–23°)

l =	68.4	71	74	157	386	35 cm.
$2r$ =	.3	.7	1.7	.7	.7	1.7 cm.
t =	15^m	15^m	15^m	15^m	400^m	10^m min.
a =	1.009	1.009	1.008	1.010	1.002	1.045 cm./sec.

The divergences in a are certainly no larger than the different errors of measurement. So far as I see, therefore, it does not seem possible to reach the relatively low value ($a = .81$) usually quoted for the diffusion of hydrogen into air by the present method.

* Advance note from a Report to the Carnegie Inst. of Washington, D. C.

** These PROCEEDINGS, **10**, 1924, p. 153.

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*WHITE LIGHT INTERFERENCE FRINGES WITH A THICK GLASS PLATE IN ONE PATH**

BY W. N. BIRCHBY

CALIFORNIA INSTITUTE OF TECHNOLOGY

Communicated, September 10, 1924

The object of this paper is to describe and account for certain interference phenomena which are observed with white light, when a thick plate of glass, or other dispersive medium, is placed in the path of one of the beams in the Michelson interferometer. The mirrors are adjusted in sodium light for circular fringes, and the paths made equal. The glass plate is then inserted in one of the paths, and that path is shortened, or the other path lengthened, by moving the mirror, till the sharpest sodium maximum is reached. The path containing the glass plate will be shorter than the other by about one-half the thickness of the plate. If white light is now substituted, the fringes should be in the field. It is better before throwing on the white light, to re-adjust the mirrors for perfect parallelism of the two beams, which may be known by the almost startling distinctness of the sodium fringes in this position. It will be easier if the first trials are made with a plate not more than 5 mm. thick.

With a thick glass plate in one path, the fringes are not all in the field at one time. It is necessary to vary the adjustable path by a millimeter or more, depending on the thickness and dispersion of the glass, in order to run through the range. Toward one end of the range the fringes are alternate circular bands of dark green and bright red. Toward the other end, the bands are bright green and dark red. In the middle of the range the bands are almost colorless, the bright circles being yellowish, and the dark ones a bluish gray. There is no determinate central fringe; the changes in coloration are so gradual that no difference can be seen in passing over even as many as a hundred fringes if the glass is fairly thick. The following table gives the results of a few trials in counting the fringes with different thicknesses of glass. The figures are not strictly comparable among themselves, as the plates were not all of the same kind of glass.

Thickness of glass plate in mm.	0	3.18	7.44	15.04	30.08
Number of fringes counted	20	255	555	1490	2500

The bands are so uniform as to suggest monochromatic light, though alternate bands are almost complementary in color. With 7.44 mm. of glass in one path, the distances passed over in counting 100 fringes at each extreme were measured, and the equivalent wave-lengths calculated. These were 6360Å and 5180Å for the bright red and the bright green ends, respectively. These are merely rough measurements, made to ascertain the general nature of the fringes. In counting fringes in these experiments, the adjustable path was slowly lengthened (or shortened) and the number of fringes vanishing (or forming) at the center counted.

The fact that interference effects are still visible, in spite of the great dispersive action of a thick glass plate, is most unexpected, and makes an explanation a matter of particular interest. The theory is given for plane wave-fronts, and is divided into two parts: I. Wave-front parallel to the mirrors. II. Wave-front oblique to the mirrors. The present paper deals with the first part. A paper covering the second part is in preparation. It should be noticed that the first part does not deal with the whole field of view in the interferometer, but only with the central spot of the fringe system.

Physical Explanation of the Fringes.—To analyze the phenomena, let us suppose at first a source of monochromatic light. The phase-difference of

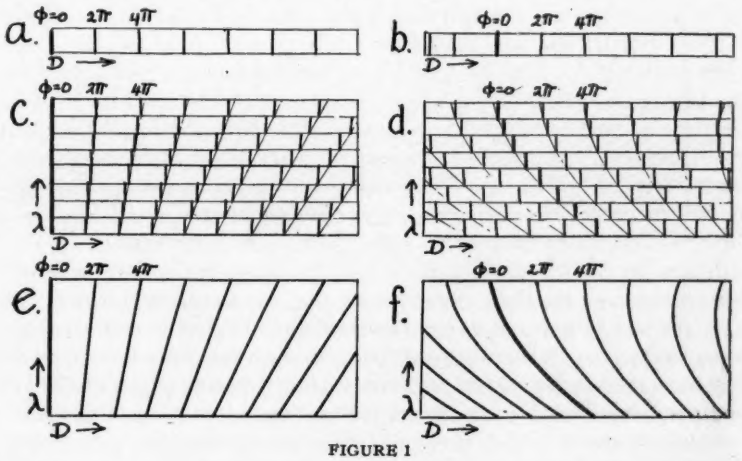

FIGURE 1

the two re-uniting beams is proportional to their path-difference, so that, if we have a field over which the difference of path increases steadily from left to right, the points where the phase-difference is an even multiple of π

(greatest light intensity) will be a series of equidistant vertical straight lines, the distance between them being proportional to the wave-length of the light employed (fig. 1A). If, now, we place a retarding medium in part of the shorter of the two paths, it will be necessary, if we wish to restore the original phase-difference, to lengthen the longer path. That is to say, any particular phase-line in the diagram will be moved to the right. The effect will be to move the system of lines as a whole to the right by an amount depending on the retarding effect of the medium (fig. 1B).

Now, if we suppose a source of light of a definite number of discrete wave-lengths, we shall have a diagram like the above for each wave-length. We can graph these one under the other as in figure 1C. Introducing the retardng medium will move each system to the right by a different amount, giving an effect shown in figure 1D, where the heavy lines are the positions of zero phase-difference in each case. Passing to the case of a light-source containing all wave-lengths, the systems of short lines merge into continu-

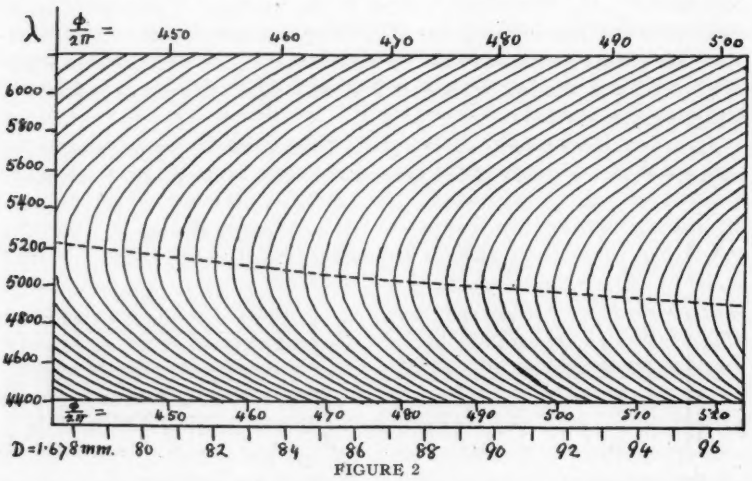

FIGURE 2

ous curves, and the phase-difference diagrams appear as in figures 1E and 1F, the former having no retarding medium. The effect of putting in a greater thickness of retarding medium is to warp the phase-lines more and more, so that we get the effect shown in figure 2, in which the point of zero path-difference is far to the left of the part drawn. The last mentioned diagram is drawn to scale, to represent the actual conditions when a glass plate 3 mm. thick is interposed in the path of one of the beams.

To explain the white-light interference effects observed when the spectrum fringes are in this condition, divide the diagram into horizontal strips of finite but narrow width $d\lambda$. This is equivalent to considering the source

of white light as being composed of a number of sources, each emitting light of all wave-lengths over a narrow range $d\lambda$. If we observe the light from one of these sources, excluding all the others, there will be visible interference if, for a given path-difference, the phase-difference does not vary much over the range. If the phase-difference at one edge of the strip differs by as much as π from that at the other edge, varying continuously and gradually from one edge to the other, the variations in intensity over the strip will compensate each other, and the field will appear uniform to the eye, since there are no decided color-differences in the small range of λ over the strip. It will be seen that this last condition holds, for a constant path-difference and a small range of λ, over the whole field in figure 2, except for those strips in the neighborhood of the bend of the phase-curves. In this region each strip will show clear interference, since the phase-differences on its upper and lower edges are nearly the same. Moreover, the neighboring strips will reinforce the interference, their phase-differences being in step with it. Over the rest of the field the action of neighboring strips will produce no interference effect, giving uniform illumination. We shall have, then, interference in one narrow range of the spectrum, superimposed on uniform illumination from the rest of the spectrum. The color effects observed can be explained if we consider the field to be composed of a set of interference bands of light of a certain color (small color-range), seen against a background of uniform illumination of all other colors. The background will be complementary to the interference bands. The intensity of the interfering color ranges from twice the intensity of the other colors to zero, since the rest of the spectrum is covered by very narrow spectrum fringes, cutting down its intensity one-half. When the interfering light is in the red, for instance, we have alternately a surplus of red and no red, similarly for other positions. The part of the phase-curves drawn in figure 2 represents less than one-third of the range of D over which fringes are visible. If the entire range were drawn it would be noticed that the position of the bend in the phase-curves passes in succession through all values of λ, from about 6000 Å. at the left, to about 5000 Å. at the right. A study of the equations given in the next section brings out this fact, which is also in accord with experiment. The dotted line shows the change in wave-length of the fringe-producing light as D varies.

It will be noticed that, according to the above analysis, the interference is caused by light of such a wave-length that its phase-difference is greater than that of any other wave-length, for the given position of the mirrors. This is in accordance with the principle enunciated by Cornu,** and enlarged upon and exemplified by Lord Rayleigh,*** that the condition for white-light interference is that the phase-difference shall be a maximum or a minimum with respect to the wave-length. In symbols, $d\phi/d\lambda = 0$, where ϕ is the phase-difference of the two beams.

Lord Rayleigh gives several methods of increasing the number of white-light fringes, by means of optical systems which tend to equalize more or less the widths of the monochromatic fringes of all wave-lengths. In the case considered in this paper no such explanation of the increased number of fringes is possible, since the "width of a fringe" is simply the distance through which the mirror must be moved to change the phase-difference by 2π, and is equal to half a wave-length in each case. Hence all the monochromatic components have fringe-widths proportional to their wave-lengths.

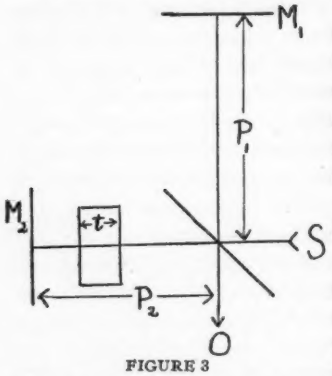

FIGURE 3

Theory of the Fringes.—In this section we are going to investigate by what distance the mirror can be displaced without destroying the interference fringes, and we shall find an expression for the number of fringes visible with plates of different thickness and dispersive power.

Let N_1 be the number of waves of length λ (in air) in P_1, (fig. 3), and N_2 the number in P_2, P_1 and P_2 being the distances from a point on the half-silvered surface to the two mirrors. Suppose a plate of glass, or other dispersive substance, of thickness t to be placed in P_2. Then, if μ is the index of refraction from air into the plate for wave-length λ, we have the relations,

$$N_1 = P_1/\lambda;\ N_2 = (P_2 - t)/\lambda + \mu t/\lambda \qquad (1)$$

Let D be the difference of the distances to the two mirrors, $P_1 - P_2$, and let ϕ be the phase-difference of the beams on re-uniting. Then $\phi = 4\pi\ (N_1 - N_2)$, since the beams pass twice over their respective paths. Directly from these relations we obtain the fundamental equation

$$\phi/4\pi\lambda = D - (\mu - 1)t \qquad (2)$$

To apply Cornu's principle, differentiate (2) with respect to λ, and put $d\phi/d\lambda = 0$ in the result. We get

$$\phi/4\pi + t\,d\mu/d\lambda = 0 \qquad (3)$$

as the condition for producing fringes in the neighborhood of λ. Eliminating ϕ between this equation and (2), we get the condition in terms of D:

$$D = (\mu - 1 - \lambda\,d\mu/d\lambda)t \qquad (4)$$

If λ_1 and λ_2 are the shortest and the longest wave-lengths of the range under consideration, by placing the corresponding values in the above equation and subtracting, we have

$$D_2 - D_1 = R = [\mu_{\lambda_2} - \lambda_2(d\mu/d\lambda)_{\lambda_2} - \mu_{\lambda_1} + \lambda_1(d\mu/d\lambda)_{\lambda_1}]t \qquad (5)$$

If μ is a continuous function of λ this can be written

$$R = t\int_{\lambda_2}^{\lambda_1} \lambda \frac{d^2\mu}{d\lambda^2}\, d\lambda \qquad (6)$$

The number of fringes in this range will be $(\phi_1 - \phi_2)/2\pi$, ϕ_1 and ϕ_2 being the values of ϕ in (3) corresponding to λ, and λ_2 respectively. We shall have from (3),

$$(\phi_1 - \phi_2)/2\pi = n = 2t[(d\mu/d\lambda)_{\lambda_2} - (d\mu/d\lambda)_{\lambda_1}] \qquad (7)$$

whence

$$n = 2t\int_{\lambda_2}^{\lambda_1} \frac{d^2\mu}{d\lambda^2}\, d\lambda \qquad (8)$$

By means of Cauchy's dispersion formula, $\mu = A + B/\lambda^2$, (2) gives the following relation between D, ϕ, and λ:

$$D = \phi/4\pi\lambda + (A - 1)t + Bt/\lambda^2$$

plotting D against λ, using ϕ as parameter, we get a series of curves of equal phase-difference. A vertical cross-section of this set of curves at any point will give the composition of the light for that particular value of D, or in other words, for the corresponding position of the movable mirror. Figure 2 was drawn from this equation, using the values: $A = 1.5$; $B = 57 \times 10^{-10}$; $t = 3$ mm.

Putting Cauchy's value for μ in (6) and (8), and integrating, gives

$$R = 3Bt(1/\lambda_2^2 - 1/\lambda_1^2) \qquad (10)$$

and

$$n = 4Bt(1/\lambda_2^3 - 1/\lambda_1^3) \qquad (11)$$

These results show that the range, and the number of fringes visible, are proportional to the thickness of the glass plate, and also to the dispersive power of the glass, as measured by B. By experiment it is found that the interference is lost in the glare of the background for wave-lengths much less than $5100A$, or greater than $6300A$. Using these limits, and taking $B = 57 \times 10^{-10}$, we get, from (10) and (11), for a plate 3 mm. thick, $R = .063$ mm.; $n = 227$. This agrees well with actual count, as can be seen from the table at the beginning of this paper.

The writer wishes to acknowledge the kindness and patience of Dr. Paul S. Epstein in reading the manuscript critically and making many valuable suggestions.

* A brief abstract of this paper was presented to the American Physical Society, September 1923. In the meantime Sethi (*Phys. Rev.*, Jan. 1924, pp. 69–74) observed the same phenomenon independently, though his theory is different from mine.

** *J. Phys.*, Ser. 2, **1**, p. 293 (1882).

*** *Phil. Mag.*, Ser. 5, **28**, pp. 77 and 189 (1889).

THE SEPARATION OF ISOTOPES BY THE IONIC MIGRATION METHOD

BY JAMES KENDALL AND JAMES F. WHITE

CHEMISTRY DEPARTMENT, COLUMBIA UNIVERSITY*

Communicated, August 13, 1924

A preliminary account of the application of the ionic migration method to the attempted separation of isotopes, with a description of the apparatus employed, has been given in a previous article.[1] It will suffice to repeat here that the method is based upon the fact that if isotopic ions possess significantly different mobilities, then the movement of a section containing these ions along a tube under the influence of the electric current should result in an accumulation of the faster ion in the forward portion, and of the slower ion in the rear portion of the sample. The boundaries of the section may be maintained perfectly sharp by having a still faster ion precede, and a still slower ion follow the isotopic mixture and, for obvious reasons, agar-agar gels are used throughout.

In the case of chlorine (at. wt., 35.46; isotopes Cl^{35} and Cl^{37}), a sodium chloride section 5 cm. long was placed between gels of sodium hydroxide and sodium acetate and moved a distance of 1000–2000 cm. before segmentation. Four runs were carried to completion. While this work was under way, the method was tested out by making shorter runs with non-isotopic mixtures of salts with ions of slightly different mobilities. By narrowing down the difference in successive experiments it was hoped to establish the lower limit at which any marked separation might be effected. The results obtained with the following typical pairs may first be cited.[2] It should be noted that comparable ionic mobility data for the concentrations used were seldom available, so that the percentage differences given are only approximate.[3]

I. *Iodide-Thiocyanate.*—A mixture of potassium iodide and potassium thiocyanate (each 0.05 *N*) was taken, with hydroxyl ion proceding and acetate ion following as before. Two runs were made; in the first the section was moved 150 cm., in the second only 40 cm. (to observe the rate at which separation was progressing). At the end of each run the mixture was divided up and the usual qualitative tests (ferric chloride and carbon disulphide—chlorine water) applied to each segment.

SEGMENT NUMBER		1	2	3	4
Run I.	Iodide coloration	Strong	Medium	None	
	Thiocyanate coloration	None	Faint	Strong	
Run II.	Iodide coloration	Strong	Medium	Faint	None
	Thiocyanate coloration	Faint	Medium	Strong	Strong

The segments are numbered from the forward end, and it will be seen

that the iodide ion, the mobility of which is approximately 16 per cent greater than that of the thiocyanate ion, rapidly concentrates in the first portions, while the thiocyanate falls back to the rear. A complete separation is here very rapidly attained.

II. *Barium-Calcium.*—The difference in ionic mobilities for this pair is about 8 per cent, the barium ion being the faster. A 6 cm. section was moved along the tube a distance of 200 cm., with potassium ion preceding and cupric ion following. Spectroscopic tests were made upon each segment and the extent of separation determined by locating the characteristic lines for the two metals.

SEGMENT NUMBER	1–3	4–8	9–11
Barium lines	Strong	Less and less persistent	Absent
Calcium lines	Absent	Increasingly prominent	Strong

Here again it is evident that a perfect separation may be easily effected.

III. *Barium-Strontium.*—For this pair the difference in mobilities is only about 5 per cent in favor of the barium ion. A 5 cm. section was moved a distance of 100 cm., and spectroscopic tests applied to the various segments as before. The length of run had clearly not been sufficient to induce complete separation, since all of the segments showed lines from both metals. The proportions present, however, changed progressively along the section, as indicated by the predominant color of the flame. In segments 1–5 this was green, in segments 6–7 red. Only a partial separation, therefore, was obtained in this case.

IV. *Iodide-Chloride.*—Here the mobility difference scarcely exceeds 1 per cent, the recorded values for 0.1 N concentration being $I^- = 57.3$ and $Cl^- = 56.5$. After a run a distance of 900 cm., the sample was segmented. Each segment was incinerated in a platinum crucible and analyzed by the Volhard method. The following table shows the number of cc. of standard silver nitrate solution required to precipitate 1 g. of silver halide from the several residues.

SEGMENT NUMBER	1	2	3	4	5
Cc. $AgNO_3$	105.2	135.1	147.6	152.1	155.6

With the same silver nitrate solution, the corresponding numbers for pure sodium chloride and sodium iodide were 104.8 and 171.3, respectively. A very considerable (although not complete) separation has consequently been induced.

These experiments, then, show conclusively that if there is any material difference in the mobility of isotopic ions (such as the difference of 1.7 per cent calculated by Lindemann[4] between Cl^{35} and Cl^{37}) the method employed is certain to give at least a partial separation. With this point established, analytical work was commenced upon the samples obtained from the four chloride runs to which reference was made above.

The results so far obtained are entirely indecisive. It is an inherent defect of the ionic migration methods (unless exceedingly wide tubes, say one foot in diameter, are employed) that only very small quantities of material are present in each segment. Not only would concentrated solutions (especially of hydroxide, the only convenient preceding ion in the case of chloride) act upon the agar-agar adversely, but it would also be impossible to keep the boundaries moving sufficiently rapidly to prevent diffusion effects from obscuring any separation obtained. Now with the tubes of $1^1/_2$ inch internal diameter used in the present work, and with sodium chloride of 0.1 *N* concentration, a segment 1 cm. thick contains sufficient salt to precipitate only about 0.1 g. of silver chloride. The evidence for isotopic separation depends upon the accuracy of the ratio Ag/AgCl, which varies only by 14 parts in 1000 for Cl^{35} and Cl^{37}. To establish a complete separation by ordinary analytical procedure would therefore be a matter of extreme difficulty, while a partial separation would be absolutely concealed under unavoidable experimental errors.

From the deadlock thus reached, several means of escape are possible. In the first place, micro-analysis might be attempted (a balance weighing to 0.001 mg. being available), but this would involve a tremendous amount of preliminary work to ensure the necessary accuracy in technique. Alternatively, the runs already made might be repeated with tubes of much larger diameter, although here again the labor required would be great. A more attractive solution of the difficulty (for the first suggestion of which we thank Professor James F. Norris of Massachusetts Institute of Technology) would be to set up a mass-spectrograph and use it to analyze the segments already obtained. A complete separation would be indicated by the absence of one of the two chlorine lines on the photographic plate, a considerable partial separation by a change in their relative intensities. It is unfortunate that this method of analysis is in no sense quantitative,[5] so that a slight separation (such as has been obtained by Harkins and others by different methods) would be certain to lead to a negative report. Nevertheless the construction of a mass-spectrograph has been started, and the samples on hand will be tested upon it as soon as it is in working order.

In the case of lead, which possesses radioactive isotopes, a still simpler test is available. A sample of a lead salt may be activated by addition of radium-D or thorium-B and the distribution of the radioactive component throughout the length of the sample determined at intervals during the actual course of the run.[6] This experiment is also in course of preparation.

To avoid misconception, it may be noted here that all this work is being performed with a clear realization of the fact that it is extremely unlikely that isotopic ions will exhibit any marked difference in mobilities, and that any definite results finally recorded will probably be negative. Nevertheless, as has been stated before,[7] an experimental demonstration of the essen-

tion equality of ionic mobilities of isotopes will be, in some respects, just as important as their actual separation. From the most recent theoretical work on the topic,[8] the only reasonable deduction appears to be that the mass of the ion does not enter into the mobility equation at all, and that Lindemann's calculations,[4] in which this factor is taken into account, are quite invalid. Direct experimental verification of this conclusion, however, is still lacking.

* Contribution No. 458.

[1] Kendall and Crittenden, *Proc. Nat. Acad. Sci.*, **9**, 1923 (75).

[2] Positive results obtained with rare earth mixtures by Dr. Beverly L. Clarke will be described in a later article.

[3] Landolt-Börnstein, "Tabellen," 5th Edition, 1923 (1105).

[4] Lindemann, *Proc. Roy. Soc. London*, **99A**, 1921 (102).

[5] Aston, "Isotopes," 1922 (114).

[6] Compare Groh and Hevesy, *Ann. Physik*, **63**, 1920 (85).

[7] Reference 1, p. 77.

[8] Born, *Zeitschrift f. Physik*, **1**, 1920 (221); Schmick, *Ibid.*, **24**, 1924 (56).

ON THE POSSIBILITY OF IDENTIFYING CHEMICAL PROCESSES IN LIVING MATTER

BY W. J. CROZIER

ZOÖLOGICAL LABORATORY, RUTGERS UNIVERSITY; AND THE CARNEGIE INSTITUTION OF WASHINGTON

Communicated, October 7, 1924

Work of recent years has shown that typical vital processes obey quantitatively the laws of ordinary chemical dynamics. Examples are found especially in publications by Osterhout and by Hecht. The demonstration of this principle was possible only when the velocities of organic activites were measured, and treated as presenting problems in mass action kinetics. In this way, as Loeb and Arrhenius foresaw and in a measure illustrated, it is possible to get around the otherwise insuperable obstacle arising from the fact that the quantities of reacting substances controlling protoplasmic activity may be extraordinarily minute, inaccessible; and that gross analysis is in any case impossible while the material is alive. These difficulties are especially conspicuous if one contemplates the investigation of so delicate a matter as the adjustor functions of the central nervous system.

Even granted, however, that equations describing the progress of change in a definable chemical system also describe effectively the kinetics of a given protoplasmic activity, we are not thereby enabled to specify the type of chemical change which may be involved, nor the sorts of substances concerned. The kinetic equations are general. A very great advance would

be obtained should it become possible for one to say, on the basis of experiments involving no destructive or other irreversible changes in the protoplasm, that a particular activity is dependent, for example, upon the velocity of protoplasmic respiration; and that another definite process is controlled by a hydrogen ion catalysis, and the like. It is proposed to describe a method which gives promise of accomplishing this purpose.

The velocities of chemical transformations obey accurately the equation proposed by Arrhenius (1889) as describing the relation between temperature and reaction velocity. This equation has now received theoretical justification by a number of workers. It states that the velocity of monomolecular change is proportional to the exponential of $-E/RT$, where R is the gas constant, T the absolute temperature, and E an energy term giving the amount of heat required for the conversion of one gram molecule of the reactant from an inactive to a reactive form. The constant E, or μ, is thus characteristic of the reacting substance. The fact that many chemical changes of the same general type (e.g., reactions catalyzed by acid) yield the same value of the critical increment E would thus be meaningless, unless it be assumed that the critical increment has reference to the formation of a common catalyst. This assumption has recently been developed by F. O. Rice. It is established, also, that in simple chemical processes the magnitude of E is a function of the particular catalyst.

It is well known that the velocities of many biological processes are influenced by temperature in a manner similar to those of reactions in general chemistry. In seeking to utilize these temperature relations with greater precision than had previously been attempted, I found that the values of the critical increment or *temperature characteristic* (μ) fall into definite groups. The assumption was made that in each of these groups one has to do with processes influenced by a common catalyst. Thus it turned out that for the locomotion of ants, and for the rate of creeping of a diplopod the critical increment is 12,230; and that in five other instances among arthropods where the rate of central nervous "discharge" is in all probability the governing influence, an identical value of the constant μ is encountered.

For the analysis of vital activities it seemed, then, that the critical thermal increment might assist in the classification of inner processes even though no definite identification of the chemical transformations concerned might be possible. As it happens, however, the analysis can in reality be pushed much further than this. A difficulty arises from the fact that on either side of a certain temperature (frequently 15° ±) the value of the critical increment is different. This discovery is especially significant for the understanding of the variability of protoplasmic activities and of the basis of adaptation. The fact itself is taken to signify that the velocities of vital phenomena are controlled by catenary series of reac-

tions having different critical increments, the actual velocities of the several steps being so related that a different reaction is the slow process on either side of the temperature at which shift is apparent. But a survey of practically all previously published work, and the investigation of a number of newly studied phenomena, shows definitely that in the case of physiological transformations known to be of similar type one encounters identical values of the constant μ. The derivation of significant values of μ depends upon the recognition of a change in the fundamental controlling reaction at certain temperatures, and upon a knowledge of the law of variation of velocities at constant temperatures. The experimental determinations indicate that in any process the latitude of variation at constant temperature is a constant fraction of the mean velocity. These findings are consistent with the interpretation that similar protoplasmic processes are controlled by the same catalysts, the concentrations of which in given cases vary within definite limits. One has every right to expect that among organisms not too distantly related there will be evidence of the presence of identical catalytic agents (*not* to be confused with enzymes).

This sort of interpretation would be strengthened were one able to point to a class of activities in which the biological catalyst might with some assurance be identified. Fortunately, this is possible. The critical increments definitely associated with cell respiration, in all cases for which data exist, are 11,500 cals. and 16,700 ± cals. In 20 series of experiments the maximum variation is 2 per cent. The former may safely be assigned to the action of OH ion. The latter is obtained for the action of a dehydrogenating system of the sulphyryl type, from data recently published by Quastel and Whetham; the significance and the widespread occurrence of this mechanism of respiration has been demonstrated by Hopkins and by Meyerhof. The critical increment for the oxidation of Fe^{II} to Fe^{III} is 16,140 calories. This increment is found in certain cases of respiration, and it is not excluded that the participation of iron as part of the complex respiratory catalyst determines the occurrence of both the increments 11,500 and 16,700.

Beyond this, it is found that some 19 distinct physiological processes the velocities of which have been measured by various workers over ranges of temperature, and for which one has in some instances direct evidence for dependence upon cell respiration, yield sensibly identical values of the constants μ. Whereas with processes obviously different (for example fermentation, and photosynthesis) the critical increments are unquestionably in different categories.

It is conceived, on the basis of these and other results, that the critical thermal increments of vital processes should (1) permit the classification of protoplasmic activities upon a dynamic basis, and (2) hold out the prospect of identifying reactions in protoplasm on the basis of the catalysts involved.

It is obvious that if this theory be in principle sound, there is available a device for the identification of chemical transformations in undisturbed living matter.

The evidence on which these deductions rest is detailed in papers appearing in the *Journal of General Physiology*.

CONCERNING SETS OF SEGMENTS WHICH COVER A POINT SET IN THE VITALI SENSE

BY R. L. MOORE

DEPARTMENT OF PURE MATHEMATICS, UNIVERSITY OF TEXAS

Communicated, September 22, 1924

A set of segments G will be said to cover a point set K *in the Vitali sense* if, for every point P which belongs to K and every positive number e, there exists a segment of G which contains P and is of length less than e.

In a recent paper[1] J. Splawa-Neyman has shown that if, in space of one dimension, K is a closed and bounded point set of measure zero and G is a set of segments which covers K in the Vitali sense then for every positive number e the set G contains a subject G_e such that G_e covers K and such that the sum of the lengths of all the segments of the set G_e is less than e. He indicates that Sierpinski has raised the question whether this theorem remains true if the stipulation that K be closed is omitted. I will show that this question may be answered in the negative. Consider the following example.

Example. Let the axis of abscissae be the one dimensional space in question. In this space let A and B denote the points whose abscissae are O and 1 respectively and, for every n let P_n denote the point whose abscissa is $1-1/2^n$. Let G_1 denote the infinite sequence of segments AP_1, AP_2, AP_3, . . . For each segment g of the set G_1 let t_g denote the smallest segment of G_1 which contains g as a proper subset and let A_g and B_g denote the right ends of g and of t_g respectively. Let S_g denote an infinite sequence of segments all with A_g as their common left end, the right end of the nth segment of S_g being located at a point whose distance from B_g is $A_gB_g/2^n$. Thus for each segment g of the set G_1 we have a definite sequence of segments S_g. Let G_2 denote the set of segments consisting of the segments of all the sequences S_g for all segments g belonging to G_1. Thus a segment belongs to G_2 if, and only if, it belongs to S_g for some segment g of the set G_1. This process may be continued. Thus we have a sequence of sets G_1, G_2, G_3. . . such that (a) for each segment g of the set G_n there exists a segment t_g which is the smallest segment of the set G_n which contains

g as a proper subset, (b) if A_g and B_g denote the right ends of g and of t_g respectively then, for each segment g of the set G_n there exists a definite sequence S_g of segments of the set G_{n+1} such that (1) each segment of this sequence has A_g as its left end, the right end of the mth segment of S_g being located at a point whose distance from B_g is $A_gB_g/2^m$, (2) for each segment r of the set G_{n+1} there exists a segment g of the set G_n such that r belongs to S_g.

Let G denote a set of segments such that (a) for every n, every segment of the set G_n belongs to G and (b) every segment of the set G belongs to some G_n. Let K denote the set of all points $[X]$ such that, for every n, X belongs to some segment of the set G_n. Clearly G covers K in the *Vitali* sense. But G contains so subset G^* such that G^* covers K and such that the sum of lengths of the segments of the set G^* exists (as a finite number). For suppose, on the contrary, that G does contain such a subset G^*. Then there are not more than a finite number of segments of the set G_1 belonging to G^*. Let g_1 denote the smallest segment of G_1 such that no segment of G_1 which is greater than g_1 belongs to $G^*$and let h_1 denote the smallest segment of G_1 which contains g_1 as a proper subset. Let C_1 and D_1 denote the right ends of g_1 and h_1 respectively. Let H_1 denote the set of all those segments of G_2 which lie on the interval C_1D_1. The segments of the set H_1 all have C_1 as their common left end and their right ends form a sequence of points which converges towards D_1 as a sequential limit point. Let g_2 denote the smallest segment of the set H_1 such that no segment of H which is greater than g_2 belongs to G^* and let h_2 denote the smallest segment of the set H_1 which contains g_2 as a proper subset. Let C_2 and D_2 denote the right ends of g_2 and h_2 respectively and let H_2 denote the set of all those segments of G_3 which lie on the interval C_2D_2. Let g_3 denote the smallest segment of the set H_2 such that no segment of H_2 which is greater than g_3 belongs to G^* and let h_3 denote the smallest segment of the set H_2 which contains g_3 as a proper subset. This process may be continued. Thus we have three infinite sequences of segments: $g_1,g_2,g_3,\ldots$; $h_1,h_2,h_3,\ldots$; and C_1D_1, C_2D_2, $C_3D_3,\ldots$ such that (a) for every n, g_{n+1} is the smallest segment of the set G_{n+1} having C_n as its left end and such that no segment of G_{n+1} which is greater than g_{n+1} belongs to G^* and has C_n as its left end, h_{n+1} is the smallest segment of the set G_{n+1} which contains g_{n+1} as a proper subset, and C_n and D_n are the right ends of g_n and h_n respectively. The segments $h_1,h_2,h_3,\ldots$ have in common a point X which belongs to K. But X does not belong to any segment of the sequence $g_1,g_2,g_3,\ldots$ Hence X does not belong to any segment of the set G^* and therefore G^* does not cover K. It is easy to see that K is of measure zero.

Definition. By a *simple chain of segments* is meant a set C of segments such that (a) the point set obtained by adding together the points of all the segments of C is connected, (b) there exists no point which is common to

more than two segments of C, and (c) no segment of C is a subset of any other segment of C.

THEOREM 1. *If, in space of one dimension, K is a point set of measure zero, G is a set of segments which covers K in the Vitali sense and, furthermore, there exists no positive number such that there are infinitely many segments of G of length greater than that number, then, for every positive number e, G contains a subset G_e such that G_e covers K and such that the sum of the lengths of all the segments of the set G_e is less than e and such that, furthermore, G_e consists of a countable number of non overlapping simple chains.*

Proof. Since K is of measure zero there exists a set of segments H such that (a) H covers K and (b) the sum of the lengths of all the segments of H is less than $e/2$. Since G covers K in the Vitali sense therefore it contains a subset U such that U covers K and every segment of U is a subset of some segment of the set H. But, by hypothesis, no segment of G is a subset of more than a finite number of other segments of G. It follows that U contains a subset T such that T covers K and such that no segment of T is a subset of any other segment of T. If P is any point of K there exists a segment g_{P_1} which belongs to T and contains P and which extends further to the right than any other segment of T which contains P. If there exists one or more segments of the set T which contain the right end of g_{P_1} let g_{P_2} denote that one which extends the furthest to the right. Continue this process. Thus we have a finite or countably infinite sequence S_P of segments $g_{P_1}, g_{P_2}, g_{P_3}, \ldots$ such that if g_{P_n} belongs to S_P and its right end belongs to some segment of T then $g_{P(n+1)}$ belongs to this sequence and to T and contains the right end of g_{P_n} and there is no other segment of T which contains the right end of g_{P_n} and extends as far to the right as $g_{P(n+1)}$ extends; but if g_{P_n} belongs to S_P and its right end belongs to no segment of the set T then g_{P_n} is the last segment in the sequence S_P. If g_{P_1} is the only segment of T which contains the left end of g_{P_2} then let h_{P_1} denote g_{P_1}. Otherwise of all those segments of T which contain the left end of g_{P_2} let h_{P_1} denote the one which extends the furthest to the left. Continue this process, thus obtaining a finite, or countably infinite, sequence L_P of segments $h_{P_1}, h_{P_2}, h_{P_3}, \ldots$ such that if h_{P_n} belongs to L_P and its left end belongs to some segment of T then $h_{P(n+1)}$ belongs to L_P and to T and contains the left end of h_{P_n} and there is no other segment of T which contains the left end of h_{P_n} and extends as far to the left as $h_{P(n+1)}$ extends: but if h_{P_n} belongs to L_P and its left end belongs to no segment of the set T then h_{P_n} is the last segment in the sequence L_P. Clearly the segments $g_{P_1}, g_{P_2}, \ldots$ and $h_{P_1}, h_{P_2}, \ldots$ form a finite, or countably infinite, simple chain C_P. Thus for every point P belonging to the set K we have a definite simple chain C_P, constructed in the manner indicated. For each point P of K let M_P denote the point set consisting of all points $[X]$ such that X belongs to some segment of the chain C_P. It can be shown that if Y and W are two points of K then either M_Y is identical

with M_W or it has no point in common with M_W. Hence there exists a finite, or countably infinite, set of points $P_1,P_2,P_3,\ldots$, all belonging to K, such that no two of the point sets $M_{P_1},M_{P_2},M_{P_3},\ldots$ have any point in common and such that, furthermore, every point of K belongs to some one of these point sets. Thus K is covered by the set of segments C consisting of all the segments of the chains $C_{P_1},C_{P_2},C_{P_3},\ldots$ But the sum of the lengths of the segments of C is less than e. The truth of Theorem 1 is therefore established.

THEOREM 2. *If, in space of one dimension, K is a countable point set and G is a set of segments which covers K in the Vitali sense then for every positive number e, the set G contains a subset G_e which covers K and which consists of a countable number of non overlapping simple chains, the sum of the lengths of the segments of the set G_e being less than e.*

Proof. Let $P_1,P_2,P_3,\ldots$ denote the points of the set K. For each positive integer n select, from the set G, a sequence S_n of segments g_{n_1}, $g_{n_2},g_{n_3},\ldots$, all containing P_n and such that, for every m, the length of g_{nm} is less than $1/mn$. Let G^* denote the set of all segments $[g]$ such that, for some n, g belongs to S_n. Thus a segment belongs to G^* if and only if it belongs to some sequence of the set $S_1,S_2,S_3,\ldots$ It is clear that G^* covers K in the Vitali sense and that if k is any positive number there are not more than a finite number of segments of G^* of length greater than k. It follows, by Theorem 1, that, for every positive e, K is covered by some subset of G^* such that the sum of the lengths of all the segments of this subset is less than e and such that, furthermore, this subset consists of a countable number of non overlapping simple chains.

[1] "Sur un théorème metrique concernant les ensembles fermés," *Fundamenta Mathematicae*, **5**, 1924, (328–330).

TIME CONSTANTS FOR ENGINEERING PURPOSES IN SIMPLE EXPONENTIAL TRANSIENT PHENOMENA

BY A. E. KENNELLY

HARVARD UNIVERSITY AND MASSACHUSETTS INSTITUTE OF TECHNOLOGY

Communicated August 13, 1924

Definition of a Simple Exponential Transient.—When a dynamic phenomenon, such as an electric current i, a velocity, or a temperature, changes from an initial steady value I_1 to a final steady value I_2, after the sudden application of a constant force at time $t = 0$, according to the exponential law:

$$i = I_2 + (I_1 - I_2)\epsilon^{-at} \quad \text{amperes} \quad (1)$$

where i is the current strength at time t, ϵ is the Napierian base, and α a time-reciprocal system constant, we may call the transient phenomenon of transition from I_1 to I_2 a *simple exponential transient*. Theoretically, such a transient requires infinite time for its completion. For most practical purposes, however, the transient lasts only a fraction of a second with most electric currents, or with most velocities. It may, however, last many hours in the case of the temperature of some dynamo-electric machine.

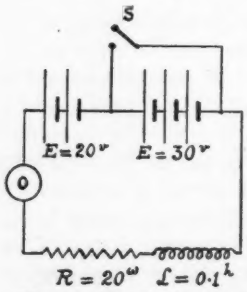

Fig. 1.—Simple circuit of exponential time constant T_ϵ = 0.005 second, decimal time constant 0.01151 second, and binary time constant 0.00347 second.

Typical Case of a Simple Electric Circuit.—Figure 1 represents the well known case of a simple electric circuit of resistance R ohms, and air-core inductance £ henries, subjected to a continuous e.m.f. from a storage battery of negligible internal resistance and inductance. A switch S, for controlling part or all of the e.m.f. in the circuit, can be suddenly opened or closed, at $t = 0$. A sensitive relay, not shown in the sketch, may be included in immediate circuit with the battery, so as to throw in an external protective resistance automatically, as soon as the switch puts the battery on short circuit. The current in the main circuit is recorded on the oscillograph O. Two well known cases present themselves, according as we decrease or increase suddenly the e.m.f. in the circuit. In the former case, with the numerical values shown,

$$i = 1{\cdot}0 + 1{\cdot}5\epsilon^{-200t} \qquad \text{amperes (2)}$$

so that closing switch S changes the current from $I_1 = 2{\cdot}5$ to $I_2 = 1{\cdot}0$ amperes. In the latter case,

$$i = 2{\cdot}5 - 1{\cdot}5\epsilon^{-200t} \qquad \text{amperes (3)}$$

or opening the switch changes the current from $I_1 = 1{\cdot}0$ to $I_2 = 2{\cdot}5$ amperes. These two cases are illustrated in figures 2 and 3 respectively.

Exponential Time Constant.—In formula (1), the reciprocal of α is known as the time constant τ of the circuit, and is equal to the ratio of £ to R

$$\tau = \frac{£}{R} = \frac{1}{\alpha} = \tau_\epsilon \qquad \text{seconds (4)}$$

so that (1) becomes

$$i = I_2 + (I_1 - I_2)\epsilon^{-\frac{t}{\tau_\epsilon}} \qquad \text{amperes (5)}$$

As other time constants will be considered, we may define this classical time constant of equation (4) as an exponential *time constant*, since $1/\tau_\epsilon$

is a coefficient of the elapsed time t, to the exponential base ϵ. An exponential time constant may be denoted by τ_ϵ. In the particular case presented in figure 1, $\tau_\epsilon = 0{\cdot}005$ second.

Exponential Transient Current-Time Curves.—In figure 2, time is marked along the axis of abscissas commencing at switch closure $t = 0$, when the current $I_1 = 2{\cdot}5$ amperes. The curve $ABCDE$ indicates the transient change of current to $I_2 = 1.0$ ampere. The rate of change of this current obtained by differentiating (1), is

$$\frac{di}{dt} = \alpha(I_2 - i)\epsilon^{-\alpha t} = \alpha\delta = \frac{\delta}{\tau_\epsilon} \qquad \frac{\text{amperes}}{\text{second}} \quad (6)$$

Where

$$\delta = I_2 - i \qquad \text{amperes} \quad (7)$$

or δ is the difference between the final current and the instantaneous current, or what may be called the *remainder*. The rate of change at any instant is therefore proportional to the remainder at that instant, and is equal to the remainder divided by the exponential time constant τ_ϵ. Ex-

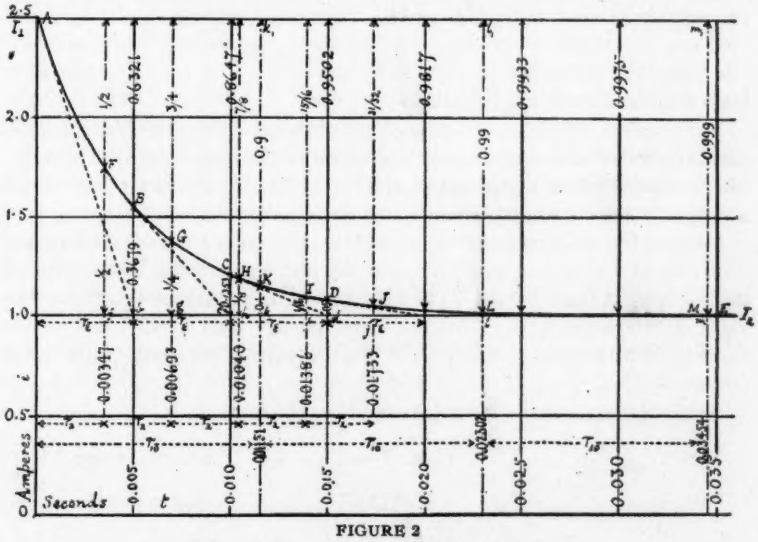

FIGURE 2

Typical descending exponential transient.

amination of the curve in figure 2, shows that the subtangent on the horizontal line I_2 of final current is constant; so that the tangent at any point say B, meets that line at a distance bc beyond, equal to τ_ϵ. This is the geometrical meaning of (6).

In one exponential time constant, the current reaches point B, which is at

a distance Bb from the final value I_2, equal to $1/\epsilon$ or ϵ^{-1}, of the total change $\Delta = I_2 - I_1$; i.e., equal to 0.3679 of that change. It has fallen $(1 - \epsilon^{-1})$, or through 0·6321 of the total change.

After the lapse of two exponential time constants, (0.010 second), the current i reaches point C on the curve which is $1/\epsilon^2$ or ϵ^{-2} of the total change to I_2; so that the remainder is 0.1353 of the change, and the *attainment* is $(1 - \epsilon^{-2})$ or 0.8647 of the change.

After the lapse of three exponential time constants (0.015 second), the *deficiency* is ϵ^{-3}, or 0.0498 of the change, while the attainment is $(1-\epsilon^{-3})$ or 0.9502 of the change. This is perhaps the easiest statistical fact to remember concerning the operation of the exponential time constant; namely, that when three of them have elapsed, the attainment towards the final state is 95.02 per cent complete, or the deficiency is almost 5 per cent.

After n exponential time constants have elapsed, the deficiency is ϵ^{-n} of the total change, or the attainment $(1 - \epsilon^{-n})$.

Similarly, at any time $t = m\tau_\epsilon$, we have a deficiency of ϵ^{-m} of the total change; so that if we know the amount of the total change, we can assign the instantaneous value, by finding the numerical value of

$$\epsilon^{-m} = 10^{-\frac{m}{2.3026}} = 10^{-0.4343m} \qquad (8)$$

from a table of common logarithms.

The simplest definition of the classical exponential time constant τ_ϵ is that time in which the transition from the initial to the final value of the variable is completed to a deficiency of ϵ^{-1}, or 36.79 per cent. This is an awkward value to remember, or explain in simple terms.

Figure 3 for the corresponding increasing case, gives a curve of transition $ABCDE$, in accordance with equation (1), and which is the same curve as that of figure 2, but inverted. The deficiency after one exponential time constant (0.005 second), is ϵ^{-1} of the whole change, and the deficiency after three such time constants is ϵ^{-3}, or very nearly 5 per cent of the total change.

Binary Time Constant.—If we write (1) in the form

$$i - I_2 = (I_1 - I_2)\epsilon^{-\frac{t}{\tau_\epsilon}} \qquad \text{ampere change (9)}$$

or

$$\delta = \Delta\,\epsilon^{-\frac{t}{\tau_\epsilon}} \qquad \text{ampere change (10)}$$

and

$$\frac{\delta}{\Delta} = \epsilon^{-\frac{t}{\tau_\epsilon}} \qquad \text{deficiency (11)}$$

where Δ is the total transition $(I_2 - I_1)$, δ/Δ is the deficiency, and $1 - \delta/\Delta$ is the attainment.

We may rewrite this deficiency as

$$\frac{\delta}{\Delta} = \epsilon^{0.69315\times-\frac{t}{0.69315\,\tau_\epsilon}} \qquad \text{deficiency (12)}$$

But $$\epsilon^{0.69315} = 2 \qquad (13)$$

so that

$$\frac{\delta}{\Delta} = 2^{-\frac{t}{0.63915\,\tau_\epsilon}} \quad \text{deficiency (14)}$$

$$= 2^{-\frac{t}{\tau_2}} \quad \text{deficiency (15)}$$

where $$\tau_2 = 0.69315\,\tau_\epsilon \quad \text{seconds (16)}$$

That is, we may transfer from the Napierian base ϵ to the base 2, by taking a new time constant τ_2, which is nearly seven tenths of the exponential time constant. We may call this time constant the *binary time constant.*

In figures 2 and 3, the first binary time constant is marked off at f, 0.00347 second from the start. At this moment, the current will have reached the point F, and the deficiency is 2^{-1}, or $1/2$, or 50 per cent. The attainment is also $(1-2^{-1})$ or $1/2$. On the lapse of two binary time constants, (0.00693 second) the deficiency is 2^{-2}, or $1/4$, and the attainment $1-2^{-2}$, or $3/4$, at point G on either curve. After three binary time constants $3\tau_2 = 0.0104$, second (a little over two exponential time constants), the deficiency is 2^{-3} or $1/8$, and the attainment $1-2^{-3}$, or $7/8$ of the whole. Similarly, on the completion of n binary time constants, the deficiency will be 2^{-n} and the attainment $1-2^{-n}$. Only five such binary time constants are marked off in figures 2 and 3, to point J, where the deficiency is 2^{-5} or $1/32$ of the total change, (0.0056 ampere).

The advantage of the binary time constant is that in this interval of time, the deficiency is 2^{-1} or $1/2$, an easy value to remember or to explain. The binary time constant does not however lend itself to exact computation at indiscriminate values of t. For such computations, the decimal time constant is preferable.

Decimal Time Constant.—We may rewrite (11) in the form

$$\frac{\delta}{\Delta} = \epsilon^{\,2.30258\times-\frac{t}{2.30258\,\tau_\epsilon}} \quad \text{deficiency (17)}$$

so that $$\frac{\delta}{\Delta} = 10^{-\frac{t}{2.30258\,\tau_\epsilon}} \quad \text{deficiency (18)}$$

$$= 10^{-\frac{t}{\tau_{10}}} \quad \text{deficiency (19)}$$

where $$\tau_{10} = 2.30258\,\tau_\epsilon \quad \text{seconds (20)}$$

That is, we may transfer to the base 10, by taking a new time constant τ_{10}, which is about 2.3 times the exponential time constant. We may call this time constant τ_{10} the *decimal time constant.*

In figures 2 and 3, the first decimal time constant is marked off at k, 0.01151 second from the start. At this moment the current has arrived at K, with a deficiency of 10^{-1}, or $1/10$ of the whole transition. The attainment is then $1-10^{-1}$ or $9/10$. On the lapse of $2\,\tau_{10}$ seconds, the deficiency is $\delta/\Delta =$

10^{-2}, or 0.01, and so on. Three decimal time constants are included in figures 2 and 3.

A disadvantage of the decimal time constant is that it is relatively so long, and the attainment is complete to within 10 per cent in its first lapse. For precise computations, however, it is very advantageous.

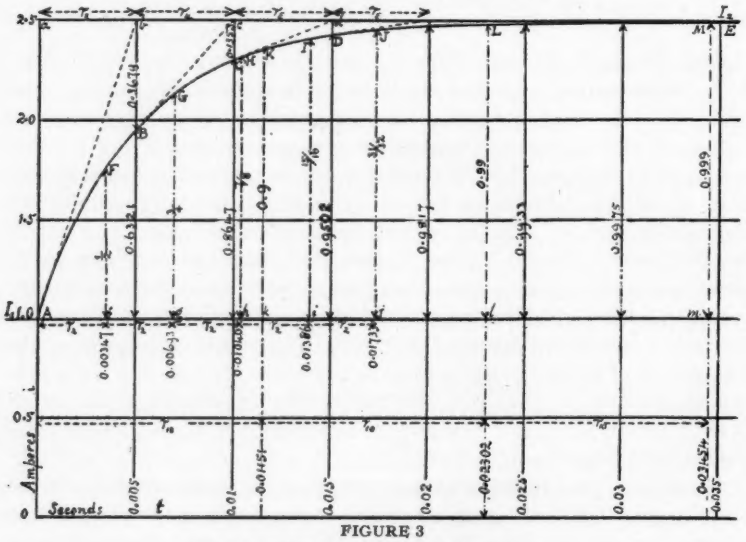

FIGURE 3

Typical ascending exponential transient.

Rectification of Exponential Transients on Arith-Log Paper.—Figure 4 shows that arith-log paper, when inverted, presents a straight line for exponential transients. The abscissas are in time constants. The ordinates are in attainments and deficiencies. Three straight lines are drawn to suit exponential, binary and decimal time constants, respectively. By the use of figure 4, the deficiency δ/Δ may be read off directly, in reference to any one of these time constants and, for many practical purposes, without further computation.

Summary.—The three time constants above discussed are all useful, the exponential τ_ϵ for fundamental theory, the decimal τ_{10} for precise computation, and the binary τ_2 for simple conception and description. Their numerical relations in all cases are:

$$\tau_\epsilon = 1{\cdot}44270\ \tau_2, = 0{\cdot}43429\ \tau_{10}$$
$$\tau_{10} = 2{\cdot}30258\ \tau_\epsilon, = 3{\cdot}32192\ \tau_2$$
$$\tau_2 = 0{\cdot}69315\ \tau_\epsilon, = 0{\cdot}30103\ \tau_{10}$$

After τ_ϵ, the deficiency is ϵ^{-1} or 0.3679, and the attainment $1-\epsilon^{-1}$.

After τ_{10}, the deficiency is 10^{-1} or $1/10$ and the attainment $1-10^{-1}$.
After τ_2, the deficiency is 2^{-1} or $1/2$ and the attainment $1-2^{-1}$.

It should be noted that the binary time-constant τ_2 corresponds to what

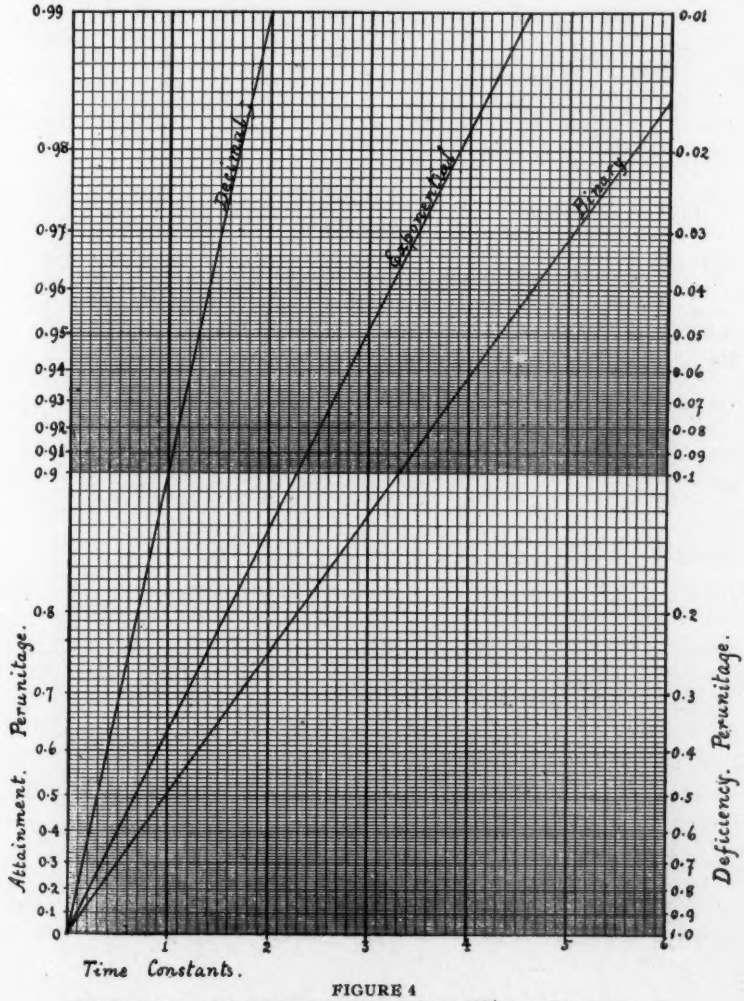

FIGURE 4
Attainments and deficiencies in terms of the different time constants.

has been known for a number of years as the "period" or "half-value period" T, in the science of radio-activity, and in the measurement of radio-active decay.